A unified theory of stable isotope fractionation driven by thermal diffusion

XUEFANG LI[1] AND YUN LIU[1,*]

[1]State Key Laboratory of Ore Deposit Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, liuyun@vip.gyig.ac.cn (* Corresponding author)

Huang *et al* (2010) found that Fe, Ca and Mg isotope fractionations of high-temperature silicate melts are only associated with the temperature gradients in thermal diffusion processes and are independent of compositions and mean temperatures [1]. Richter *et al* (2010) doubted that the existing data are sufficient to obtain such conclusion [2]. A few theoretical models have been proposed for explaining isotopic fractionations in these processes under high temperatures [3, 4]. However, molecular-level mechanisms and theoretical treatments of these processes are still under debating.

Here we provide a unified theory based on the local thermodynamic equilibrium treatment (LTE) of statistical mechanics for evaluating isotopic fractionations under thermal gradients. Our theory can be used to high- or low-temperature situations instead of only under high-temperatures.

Under high temperatures, our theory however can be reasonably approximated to this equation:

$$\Delta^{X} M = A \ln \frac{T}{T_0} + B\left(\frac{1}{T^2} - \frac{1}{T_0^2}\right)$$

where A and B are constants which are related to specific isotope systems and chemical compositions of silicate melts.

If the thermal gradient is not very large and the mean temperature is high, the second part of the above equation can be safely neglected and obtain an extremely simple equation which is linearly dependend on temperatures, agreeing with what Huang *et al* (2010) concluded.

Based on this terse equation, we can not only easily provide isotope fractionation data for almost all kinds of isotope systems, but also can provide the mechanisms of isotope fractionation in thermal diffussion processes.

[1] Huang *et al* (2010) *Nature* **464**, 396-400. [2] Richter *et al* (2010) *Nature* **472**, E1-E1. [3] Dominguez *et al* (2011) *Nature* **473**, 70-73. [4] Lacks *et al* (2012) Phys. Rev. Lett. **108**, 065901.